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$\bar{R}$	Universal gas constant
$t$	Time
$T$	Absolute temperature
$T_{Tr}$	Absolute temperature of the drop [Tr = Tropfen, drop]
$T_{\infty}$	Absolute temperature at infinitely great distance from the drop
$T_0$	Room temperature (293° K)
$T_1$	Body temperature (310° K)
$V_{fl}$	Volume which a water molecule of the liquid phase occupies ( $3 \cdot 10^{-23}$ cm) [fl = flüssig, liquid]
$V_{Gas}$	Volume of a gram mole of water vapor
$V_{fl}$	Volume of a gram mole of water in the liquid phase [fl = flüssig, liquid]
$V_{Kri}$	Volume of a salt crystal (or volume of the amorphous substance) [Kri = Kristall, crystal]
$V_{Tr}$	Volume of a salt water drop [Tr = Tropfen, drop]
$W$	Quantity of heat produced by water vapor condensation [W = Wärme, heat]
$\dot{W}$	$\frac{dW}{dt}$
$\lambda$	Thermal conductivity
$\mu$	Length of $10^{-4}$ cm

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- [Word illegible: Number?] of water molecules in drop of radius  $R$
- [Words illegible: Number of molecules?] employed in cyclic process
- Density of water
- Density of sodium chloride solution of concentration  $c$
- Surface tension of water (73 dyn/cm)
- Integral of Gaussian error function

STUDIES OF THE ELECTRICAL SPACE CHARGE DENSITY  
OF DISPERSION AEROSOLS

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pages 288-303

Dr Karl Biss, Silicosis Hospital,  
Grafschaft/Hochsauerland, and  
the Physicommedical Research Cen-  
ter (Chief Physician: Dr Biss)

The electrical charge of an aerosol is an essential cause of the characteristic behavior of the carrier substances in the gaseous phase. It influences the lifespan and limits the practical possibilities of application. The tendency of an aerosol towards homogenization, the process of aging, takes place through the growth of larger suspended particles at the expense of smaller fractions in the particle-size spectrum. Aggregation and conglomeration are controlled by the electrical charge properties of the aerosol, a phenomenon which is known as recombination of suspended particles with unlike charge sign. These phenomena by virtue of the space charge of the atmosphere are of the greatest significance for the dynamics of hydrometeors. However, in lesser orders of magnitude and ranges as well the electrical charge affords an opportunity for different manipulations on an aerosol, either for the purpose of stabilization or with the object of rapid sedimentation.

The formation of a space charge field in which the aerosol particles subject to the force of gravity give up their charges on sedimentation surfaces is produced by space charge density. In the biological process it can be assumed that the function and dynamics of cell complexes are vigorously affected by electrical phenomena, as, for example, is probably done by the permeability theory or as is also conceivable by the charge properties of protein molecules. -- Recently a number of investigations have been published, dealing with the effects of atmospheric electrical fields and unipolarly charged suspended materials on the human organism.

Possibly uncertainty as to quantitative electrical charge relationships under the experimental conditions is a reason for the (in part) significantly divergent interpretations.

Therefore, in the following discussion the causes of the electrical charges in an aerosol are presented although many explanations no longer stand up under criticism today. Thereafter a report is made on our own experiments, which had as their purpose to survey the electrical space charge density in different aerosols and which set forth in detail the magnitudes of therapeutic aerosols. I consider it justified to publish a part of these current experiments since the significantly improved methods of measurement permit comprehensive reproducibility and surpass previous methods in respect of accuracy of measurement.

#### On the Theory of the Electrical Charges of an Aerosol

Kohlschütter and Remy have explained that during quietly proceeding chemical reactions aerosols can be formed that have no electrical charge. Therefore the mechanical forces acting upon a fluid during the atomization process can be considered to be the cause of an aerosol's electrical charges. The electrical charges therefore are due to the spraying of very fine negatively charged fluid drops from the electric double layer on the free fluid surface which is always formed in the case of substances with high associative property and high dielectric constant. Accordingly, electrical space charge density is characterized by the resultant negative carriers of electricity which originate from the topmost negatively charged molecular layer.

According to Lenard, the electric double layer of a water droplet is in the air with both its plates in the droplet. Therefore in pure water the negative charge is situated on the surface and extends to a depth of  $8 \times 10^{-7}$  cm. The positive charge is situated further inside up to  $13.0 \times 10^{-7}$  cm. In the above example, therefore, the thickness of the double layer is about 40 molecular diameters. However, the capillary layer on the surface is considerably thinner.

Evidence of the charge distribution in the electric double layer is obtained by the well-known fundamental experiment in which the travel of a small air bubble in the electrical field of a capacitor filled with pure water is observed. If the ascent of the air bubble is suppressed by uniform rotation of the capacitor in the horizontal longitudinal axis, the air bubble travels to the anode. The gases appear accordingly always to be negatively charged in fluids. The charge cannot,

however, affect the gas molecules since thus far it has not been possible to discern gas ions in bubbles after contact with fluids. It can therefore be assumed that the fluid contains the negative charge carriers and the entire field of the electrical contact layer is in the topmost molecules of the fluid. In the above experiment, therefore, only one very thin-walled gas-filled fluid bubble travels and over this bubble the positive fluid ions in the subsequent molecular layers glide invisibly to the anode (Pohl, Elektrizitätslehre [Elements of Electricity]).

Thus during destruction of the electric double layer parts of the outer layer are knocked off. If the dispersed droplets are smaller than the external negative layer -- less than  $8 \times 10^{-7}$  cm, they carry a negative charge. The presence of very small positive carriers is explained by the conception that during injury to the electric double layer the positive plate is also exposed for a short while so that positive carriers can also be liberated from the destroyed surface in the event of rather powerful mechanical action. Added in behalf of this assumption is the fact that in the case of strong dispersion phenomena the quantity of positively charged carriers increases, the ratio of which to negatively charged particles is 2:30. Since the droplet diameters here cannot be differentiated significantly according to the sign of the charge, we are not dealing with any aerosol residues in the case of the positive carriers.

Phenomena during the atomization of fluids have been investigated with considerable expenditure of labor, Leonard, Christiansen, Pomeroy, Kohlshütter and Chapman being especially outstanding. The theory of waterfall or balloelectricity in the above descriptions is valid for chemically uniform fluids in the case of which the surface is the site of the electrical charge. With the addition of acids or bases, however, the charging direction can be reversed so that the concentration of the dissolved electrolyte in the surface layer and, hence, the adsorption properties take on essential significance for the formation of the electrical charge and of the sign as well. It has been established that carrier formation is closely related to the electrolyte content of the initial fluid since the formation of a strictly constructed electric double layer, as in the case of pure water, is interrupted in fluids of differently dissociated ions. To be sure, however, a number of other factors have to be considered since the results of our experiments cannot without further ado be harmonized with the previous interpretations either. Besides, the presence of an electric double layer in the case of droplets with a higher rate of evaporation under  $0.1 \mu$  in diameter is conceivable only with difficulty.

In order to understand the behavior and the formation of the electrical charge of dispersion aerosols, it seems advisable that a consideration of the physical processes and concepts come first.

Contact potential: In the case of the approach of two solid bodies, e. g. insulators at a molecular distance of the order of  $10^{-8}$  cm apart, electrical fields appear in the contact layer, which are characterized by very short lines of force. The one body gives up to the other electrons which are adsorbed in the receptor and arranged on the surface in a specific manner. Now this field corresponds to the electric double layer, while its voltage is defined as contact potential. The electric double layer represents a surface covered with dipoles. Dipoles can be conceived of as molecules with electrically polar properties which possess two electric charges of equal magnitude and opposite sign  $-Q + Q$  at interval  $\Delta s$ . Dipole moment is the result of the product of the charges and the interval. Used as the unit of measure of charge  $Q$  is an integral multiple of the smallest charge, of the electric elementary quantum  $e$ . This electronic charge has been defined as

$$e = (1.6020 \pm 0.0004) \times 10^{-19} \text{ C}_{\text{abs}}$$

by means of the U [as in text] droplet procedure (Überschichtenverfahren) in the suspension capacitor (Schwebekondensator).

In fluids and gases dipoles have the tendency in the presence of an electrical field to adjust themselves in the direction of the field. In the process they are continually disturbed by thermal motion. The magnitude of the dielectric constant increases with increasing orientation. Now the Ueborn law of charge holds true for contact potential between a solid and a fluid body. According to this law the substance with the greater dielectric constant is positively charged. Dielectric constant is essentially dependent on density. With decreasing density the constants approach a limiting value, at which the relative dielectric constant can be equated with the absolute dielectric constant of the vacuum. However, the substance contains quasielastically bound electrical charges which undergo a shift through the electrical field so that the dielectric constant of the substance is greater than the limiting value is. These shifts are the cause of the behavior of a dielectric vis-a-vis electrical fields. Consequently, the dielectric constant is determinative for the electrically fundamental properties of the dielectric.

The dielectric constant of the gaseous phase of an aerosol in the physical normal state ( $0^{\circ}\text{C}$ , 760 torr) reveals only slight deviations in comparison with a vacuum. However, a dispersed aerosol phase behaves differently, with significant differences appearing in liquids because of dielectric displacements. Orientation of the dipoles takes place in accordance with electrostatic laws. The opposed poles approach the neighboring dipole molecules. Thus, poles of like charge withdraw from one another so that the attractive influence of poles of unlike charge is considerably stronger than the repulsive effect of poles with like sign. This phenomenon of attraction and accumulation is designated dipole association. Molecules of water can also be bound to other polar molecules and groups of atoms, by hydration for example. The paraffin chains of fatty acids cannot accumulate such water molecules since they are apolar and not hydrated. As already mentioned, the orientation of the dipoles in an electrical field is disturbed by thermal agitations, with a characteristic equilibrium setting in at a specific temperature. Thus the intense temperature-sensitivity of the dielectric constant is understandable. In the event of obstruction of polar orientation, e. g. by freezing, the dielectric constant decreases significantly. Whereas water at  $18^{\circ}\text{C}$  has a dielectric constant of 81.6, in the case of ice the constant declines to a value of 3.1. In the solidified state the dipoles are of course firmly fixed in the crystal lattice.

Electrostatic processes are operative in aqueous solutions of electrolytes too. The ions try to disperse and arrange themselves, with more ions of the opposite sign gathering in the vicinity of an ion. This pseudostucture is likewise disturbed by the thermal agitations of the ions of dissolved electrolytes. Screening by space charges accordingly produces a loss in the activity of the ions in a concentrated solution since every ion is obstructed in its mobility. Consequently the osmotic, conductometric and electromotive dynamics of every ion in concentrated solutions is less than in highly dilute solutions. Electrostatic effect -- and therefore with ionic strength -- increases with valence too. In comparison with pure water aqueous electrolytes have heightened electric conductivity. The substances producing electrolytes decompose in solution into more or less charged electric radicals. The strong electrolytes are practically completely dissociated in weak solutions. On the other hand, the weak electrolytes do not completely decompose even in the smallest concentrations. The dissociation of electrolytes is all the greater, the more dilute the solution is. If the ions are considered as molecular, then the molecular concentration of an electrolyte is greater than the gram mole content of the

corresponding undissociated substance. These phenomena seem worthy of mention because the dielectric constant generally is also dependent on the concentration and the degree of dissociation of the dissolved electrolyte.

Further, the shape of the molecules must be taken into consideration for the structure of the electric double layer. Thus protein molecules appear in first approximation as ellipsoids of revolution, which on further investigation are recognized as rod-shaped molecules. From the example of the protein molecule it was possible to prove that charge conditions are different on the surface and in the interior. pH-shifts are also measurable, which from the surface to the interior can amount to as much as 0.78 pH units.

Finally, for the formation of the electrical charge of dispersion aerosols we must take into consideration a process which is known as adsorption. Here we are dealing with the concentration of substances at the phase interface. A number of forces take part in physical adsorption -- van der Waals-London dispersion forces which become operative between the dipoles, multipoles or ions. In addition, electrostatic forces develop which result in a binding of molecules to the surface. The concentration of substances in the surface proceeds according to the Gibbs law of adsorption according to which the amounts of the substance adsorbed from the gaseous phase can be ascertained by measurement of surface tension. Surface tension depends on the specific capillary activity of a substance, which is characterized by the Szyzskowski formula and Traube rule. Thus, substances which possess greater surface energy than the solvent and, accordingly, heighten the surface tension are displaced from the fluid surface. In particular, electrolytically dissociated salts are said to be surface-inactive since they are capable of raising the surface tension, e. g. of water, at most negligibly. The surface tension of a liquid is lowered, however, by a surface-active substance with less surface tension being concentrated at the interface. Since surface tension can already be affected by monomolecular layers, traces of surface-active substances readily exert an effect.

It can be discerned that the formation of electrical charges during aerosol dispersion is no simple linear function of physical phenomena, but a complex series of factors working together. It is also clear that the most simplified conceptions of waterfall electricity do not suffice for an understanding of electrical space charge density in the case of dispersion aerosols. First in an analysis of the relationships we must take into consideration the energy expended in

spraying, as well as dilution effects depending on the peculiarity of the aerosol generator. The particle-size spectrum is also dependent on these conditions. Apart from the chemical properties of the fluid, we must, for example, mention its viscosity as well as the physical state of the gaseous phase, both of which affect the aerosol. In accordance with the preceding remarks the formation of an electric double layer on the free fluid surface plays an important part. A lasting impression is left on this role by dielectric constant, the dissociation of electrolytes, by dipole character and dipole moment as well as by the form and shape of the molecules. Finally, adsorption processes have a direct effect on the formation of the electrical charge. If we assume that a number of electric double layers can be present in an aerosol droplet, the conception of the active mechanism for the formation of such carrier charges is more complicated still. Moreover, we started from the assumption that in the case of the aerosols we are dealing with rod-shaped droplets. However, in the case of turbulent process an increase in the relative movement of the drop in comparison with the surrounding air appears so that oscillatory motions occur during which the rod shape of the droplet changes into a free flow form. In the case of aerodynamic deformation of a droplet the undisturbed formation of an electric double layer is not possible, and in such case a further factor of uncertainty appears.

We can delve more meaningfully into the problems of electrical charges of dispersion aerosols by means of experimentation. With the present experiments only a partial insight can be given into electrical relationships of carrier substances, restricted to the viewpoint of therapeutic aerosols.

#### Performance of Experiments

Suspended particles in gases possess electrical properties by reason of a spatial distribution of the charge. The latter being an excess charge of one sign is susceptible of measurement. Used as unit of measure is the number of elementary charges in a volume of 1 cu cm. Electrical space charge density is measured with a device according to Muhl-eisen [See Note] (Figure 1). The apparatus consists of a highly insulated suspended filter in the intake part of blower, in which a flow velocity of 2 liters per second is produced. The suspended particles held fast on the filter give up their electrical charges via eligible series resistances in a range from  $3 \times 10^{11}$  to  $1 \times 10^6$  to a vibrating condenser measurement amplifier (FH 408). The sensitivity of

the instrument in conjunction with a resistance of  $10^{11} \Omega$  is  $10^{-15} \text{ A}$ . The measured voltage appears as a variable in the formula

$$v = \frac{U}{e \times R \times v} \quad (\text{e/cm}^3)$$

where  $U$  stands for voltage in mV,  $v$  for volume in  $\text{l/s}$  and  $e$  the electrical elementary quantum  $1.6 \times 10^{-19} \text{ C}_{\text{abs}}$ . Resistance  $R$  varied in a range from  $3 \times 10^{11}$  to  $1 \times 10^7$  ohms.

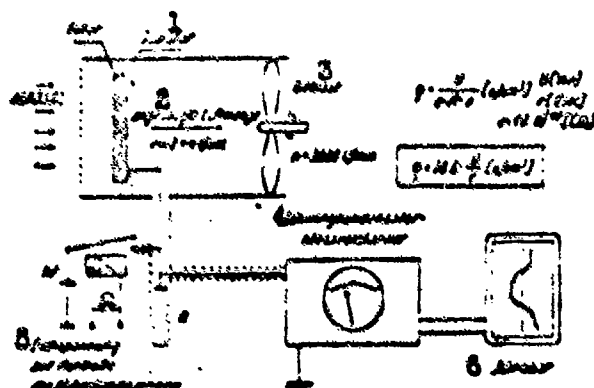


Figure 1.

Keys:

1. Insulator
2. Quantity of air sucked in
3. Blower
4. Vibrating condenser - measurement amplifier
5. Calibration voltage for monitoring resistance
6. Recorder

Measurements were made under individual experimental conditions at 10 sec intervals, with the mean value of a 4 sec observation being recorded. As aerosol generator we employed a series-produced ring nozzle (Hoyer). The atomization attachment with outlet aperture was at a distance of 20 cm from

the aerosol filter. By installing a conical shield, disturbance deflections, e. g. from people walking around, were prevented. After every experiment the atomizer, well cleaned after a washing with distilled water, was dried in the sterilizer and in each instance loaded with 4 cu cm of the fluid to be investigated. During a part of the experiments aerosol generation with oxygen as the propulsive gas was undertaken in parallel fashion, but otherwise entirely with well-cleaned compressed air. No significant differences were obtained for oxygen and air given equal propulsive pressure. Atmospheric space charge density was eliminated before each experiment by zero-point regulation of the measuring equipment. Atmospheric pressure exhibited fluctuations from 752 to 768 mm. Relative air humidity was kept constant at about 70% and an air temperature of 16° C was measured.

[Note]: At this point let me thank Decent Dr. Muhlisen, Max Planck Institute of Stratospheric Physics, for his expert advice.

Experiment I: The purpose was to establish the extent to which variable propulsive pressure of the aerosol generator affects the formation of electrical space charge density. From Figure 2, in which the experimental results are recorded, it can be seen that with increasing mechanical action on the fluid and with a heightening of dispersion forces the number of elementary charges decreases correspondingly, and in the case of an aqueous aerosol the excess of negative charges rises significantly. A relatively flat course of the curve is discernible in a pressure range from about 1.2 to 1.8 atmospheres absolute pressure. Therefore in all experiments we chose a propulsive pressure of about 1.6 atmospheres absolute pressure, in which range minor pressure fluctuations caused only slight breadth of error of the charge balance.

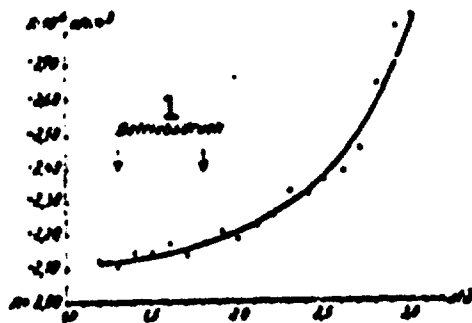


Figure 2.

Key: 1. Operating pressure

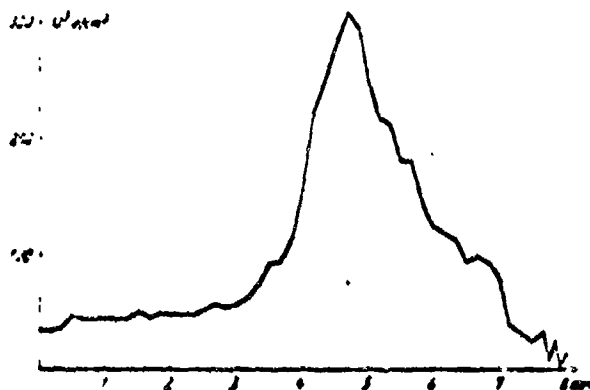


Figure 3.

**Experiment II:** The purpose was to ascertain the electrical space charge density of an aerosol of distilled water in relation to the length of atomization. Results are recorded in Figure 3. About three minutes after approximately constant behavior of space charge density a steep rise in negative charge balance becomes evident, which inside eight minutes drops to a minimum with severe fluctuations. These phenomena are no doubt caused by atomization processes. Inside the first few minutes the supply of fluid is uniformly used up. Thereafter large-bubble atomization sets in with bursting of the fluid residues in the aerosol container and of the returning fluid drops. The violent fluctuation of the charge balance can thus freely be brought into correlation with inadequate atomization. Three minutes later aerosol concentration was considerably less. Therefore, for subsequent experiments only measurements within the first three minutes were usable.

**Experiment III:** Soda lye, ammonia, common salt, hydrochloric acid, sulfuric acid and acetic acid were atomized. We dealt with normal solutions ranging fractionally from 1 n to n/10,000. The arithmetic mean of the charge balance was taken and recorded for every individual experimental condition as in Figure 4. These experiments were repeated in their totality at a later time with far-reaching reproducibility being obtained. Discernible as a significant result is the fact that acids and lyes behave in the opposite sense as regards the sign of the electrical space charge density in the event of a differing degree of dissociation. The acids employed

have a negative sign at a higher electrolyte concentration and change over to a positive charge surplus at  $n/100$  to  $n/1000$ . Common salt also behaves similarly, while soda lye from the positive range yields a negative charge balance in the event of a higher dilution. On the other hand, ammonia remains unaffected in the negative range from  $1/n$  to  $n/10,000$ . The magnitude of the charge excess is clearest given a high concentration and high dilutions.

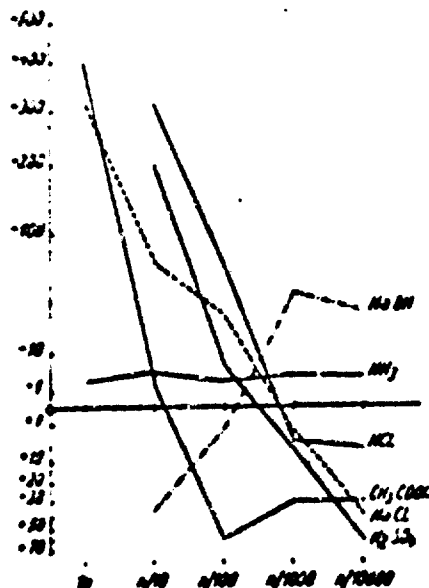


Figure 4.

Discussion of the experiment: Normal solutions of acids of higher concentration tend towards a negative charge balance, which changes the charging direction with increasing dilution. Lyes, on the other hand, exhibit the opposite behavior in that they lose their positive charge excess with increasing dilution when they are converted into the aerosol state. Weak electrolytes, as ammonia for example, have a smaller negative electrical space charge density, which undergoes no significant change with increasing dilution. Concentrated solutions of common salt have a negative charge excess; there is no change-over until the dilution is  $n/1000$ . These experiments were continued for a series of additional lyes and acids, which owing to their compatible behavior in another connection are to be presented separately.

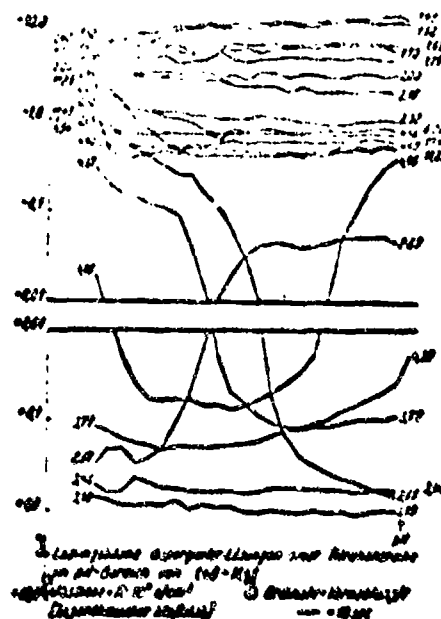


Figure 5.

**Keys:**

1. Charge balance of dispersed solutions of a titration series in the pH range from 1.48 to 11.48
2. Abscissa =  $X \cdot 10^4 \text{ e/cm}^3$   
(logarithmic scale)
3. Ordinate atomization/%  
— = 10 sec.

**Experiment IV:** In this series of experiments the purpose was to check the electrical charge of sprayed solutions in the pH range from 1.48 to 11.48. The titration series was prepared with n/10 normal solutions of soda lye, hydrochloric acid and acetic acid according to Table 1. Experimental set-up and scope of the measured values were as in the fundamental experiment.

Table 1

NaOH n/10	HCl n/10	CH <sub>3</sub> COOH n/10	pH value
0	10	10	1.43
1	9	10	1.52
2	8	10	1.57
3	7	10	1.63
4	6	10	1.70
5	5	10	1.77
6	4	10	1.87
7	3	10	2.00
8	2	10	2.15
9	1	10	2.32
9.5	0.5	10	2.55
10	0	10	3.10
10.5	0	9.5	3.45
11	0	9	3.77
12	0	8	4.13
13	0	7	4.37
14	0	6	4.50
15	0	5	4.73
16	0	4	4.80
17	0	3	4.90
18	0	2	4.91
19	0	1	5.00
19.5	0	0.5	5.00
20.0	0	0	5.37
20.5	0	0	11.23
21.0	0	0	11.48

Points of equivalence of a titration series. A stronger acid is first neutralized with the addition of n/10 NaOH to the acid mixture. Binding of acetic acid with the formation of sodium acetate does not occur until all the hydrochloric acid is saturated with simultaneous formation of the neutral salt. The resulting points of equivalence are calculated approximately and confirmed by measurement.

Discussion of the experiments: The results recorded in Figure 5 show considerable fluctuation of the charge balance in the range of pH 2.65 and pH 4.37. With increasing length of atomization the space charge densities of these sprayed solutions change over from the negative range to the positive range. The opposite relationships are revealed in the case of solutions with a pH value of 5.67 and less clearly

in the case of solutions with a pH of 4.15. Solutions with the pH values 3.77 and 3.46 and 3.10 show a relatively constant positive charge balance, while all the other solutions employed have a higher negative electrical space charge density. It follows from Table 1 which quantities were added gradually to the acid mixture in order to neutralize the stronger acid first with simultaneous formation of neutral salt. Only after this does binding of the acetic acid take place with the formation of sodium acetate. With progressing neutralization, attainment of the point of equivalence makes itself known by a jump in the pH value. The first jump occurs during the neutralization of hydrochloric acid with a pH value of 3.10. Characteristically, solutions with a pH value of 3.10 and 3.77 attain a positive charge balance. The variable space charge density and the change of sign of the test solutions with pH values 4.37 and 4.15 can possibly be correlated with the degree of dissociation of the electrolyte and with an unstable rearrangement of the atomized droplets in the electric double layer. Changes in concentration probably also explain the special behavior of the solution with pH value 2.65, which in the vicinity of the first point of equivalence assumes a positive charge excess with increasing length of atomization. The behavior of the other solutions up to a pH value of 6.00 is produced by the dominant effect of the acetic acid. Less understandable is the behavior of the solutions with pH values 11.23 and 11.48, which have a slight negative charge excess. This, to be sure, can be caused by the proximity of the second point of equivalence and of the pH jump, something to which the changing charging direction of pH solution 8.87 from the positive to the negative range attests.

During neutralization of the stronger acid the pH solutions in the neighborhood of the point of equivalence become labile in their charging direction and in charge intensity. Also the point of equivalence of the weaker acid can be well observed within the values for space charge density of the atomized solutions. By virtue of the variable (in relation to the length of atomization) charge balance, which is revealed especially drastically in the case of the different solutions, the probability exists of a defective orientation of the molecules in the droplet surface and of an incomplete formation or transformation of the electric double layer in the aerosol droplet. Hydrogen ion concentration alone is of no essential significance for electric charge balance, but it makes itself clearly evident in the neighborhood of the points of equivalence depending on the duration of atomization.

Experiment 7: The electrical space charge density of a number of inhalation drugs was tested, adhering to the dilutions indicated by the manufacturers for aerosol therapy. The

experimental procedure corresponded to that of the fundamental experiment. The measured values obtained at 10 sec intervals were recorded for three minutes and the arithmetic mean taken. We can forego indication of the scattering range since the values within one experiment revealed no fluctuations worth mentioning. The results are presented in Table 2.

Table 2

Drug	Charge balance
Kamochin	$1.25 \times 10^6$ e/cm <sup>3</sup>
Contobon	- 2.69
Aludon	- 3.12
Tyromolvin	- 3.13
Proxylmethyl	- 5.17
Oxyprocan	- 5.52
Transphidrin-Ephedrin	- 5.52 -- $\pm 0$
Euphyllin	- 7.64
Prictan IV	- 10.70
Oxyprocan-Brox	- 12.13
Prictan	- 13.50
Repanthion	- 18.05
Pyoniva	- 19.50
Aristamid	- 24.61
Serpanil	- 25.82
Solu-Supronal	- 28.41
Nephulen	- 29.62
Streptomycin	- 31.64
Inhalopen	- 33.21
Penicillin-Aerosolat	- 34.36
Lokastin	- 35.12
Paraben	- 35.84
Periston	- 36.51
Aspasan	- 40.43
Brox	- 46.78

Atomization data: Hoyer ring nozzle; propulsive pressure 1.6 atmospheres absolute pressure oxygen.  
Aerosol temperature  $16.8^{\circ}$  C.

Discussion of the experiment: Essentially a negative charge balance is revealed by the several therapeutically common aerosols here investigated. Electrical space charge density ranges from 1.25 to  $-46.78 \times 10^6$  e/cm<sup>3</sup> and is extraordinarily high. Kamochin showed a moderate positive charge,

while only Transpulmin-Sphedrin tended to the zero point, to a charge equilibrium. If we take the usual inhalation time during a single inhalation as the basis, we obtain hence the amount of excess charges which can be calculated with a multiplier of  $1 - 2 \times 10^4$  and the measured electrical space charge density of the aerosol per cu cm. Hence we freely gain the idea that, apart from the pharmacological action of a therapeutic aerosol, the charge emission on the resorption surface cannot by reason of its magnitude be a matter of indifference.

On the basis of the above results the question arises of the electrical space charge density of electro-aerosols in a space aerosol.

Experiment VI: The measuring apparatus was about two meters away from an electro-space aerostat. The aerosol generator and the charge system were set in motion at the same time. Two ultrasonic atomizers were employed as aerosol generators, and their aerosol ( $\text{NaCl } 3\%$ ) conducted into the charge system, on which a voltage of 60 kV had been applied. Measurements could not be made regularly due to the technical set-up of the experiment, but are recorded in time and presented in Figure 6. After the aeration system is turned on, at a high aeration rate (2 cu m/min) only a slight shift of the small negative charge excess to the positive range is revealed. After the aeration is turned off, the negative charge excess rises steeply and is relatively stable during the rest of the experiment. Owing to the voltage applied to the charge system, an analogous-sign unipolar charge and charge-exchange of the aerosol was produced. The steeply rising space charge density after the aeration was turned off proves that, given this set-up, space charge effects are caused essentially by the expanding, unipolarly charged aerosol and cannot be due to field action between the voltage-conducting parts and earth potential. During these experiments electric space charge density was measured at a maximum of  $160 \times 10^3 \text{ e cm}^3$ , corresponding to  $0.16 \times 10^6 \text{ e/ccm}$ . The electric charge balance in the case of an electro-aerosol under the above-mentioned conditions is relatively small. In the above-described experimental set-up it is far from reaching the electrical space charge density of therapeutic aerosols in the individual atomizer.

A report will be made in the near future on electric charge balance in the biological sphere during inhalation of dusts and aerosols.

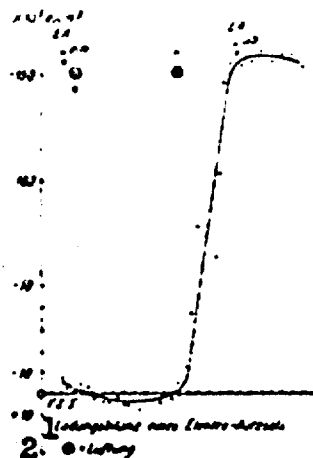


Figure 6.

Keys:

- EA: Electro-aerosol  
 ein: on  
 ab: off  
 1. Charge balance of an electro-aerosol  
 2. Aeration

Summary

The causes for the formation of the electrical space charge density of dispersion aerosols are discussed. The essential formation of a more or less strictly constructed electric double layer in the fluid drop is linked with a number of factors, among which electrolytic dissociation, dipole moment and dielectric constant, the form of the molecules and adsorption phenomena etc. must be considered. In addition, the energy consumed in atomization as well as particle size and aerosol concentration play a significant part. Electrical space charge density can be measured relatively exactly exactly with the filter method according to Muhleisen. By means of this procedure an increase in electrical space charge density with increasing jet-propulsion pressure was ascertained. Charge balance is relatively constant depending on the duration of atomization. Only with a shortage of fluid does large-bubble and irregular atomization appear, which results in steeply rising space charge density. Acids have a nega-

tive charge sign in the event of concentrated electrolytic content, whereas in the case of greater dilutions they have a positive charge excess. Common salt behaves similarly, while lyes of the positive region enter into a negative charge balance with increasing dilution. Solutions of a titration series in the pH range from 1.43 to 11.48 reveal a labile charge balance with a partial change of sign insofar as they and their pH value are in the vicinity of the points of equivalence. A number of aerosol drugs have a negative charge excess, which was measured as essentially negative up to  $46.78 \times 10^6$  ecm. On the other hand, a check of an electro-aerosol under specific conditions adduced only an electrical space charge density of  $0.16 \times 10^6$  e/cm<sup>3</sup> so that the electrical charges released during inhalation of a therapeutic aerosol are by no means insignificant.